

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
21 March 2002 (21.03.2002)

PCT

(10) International Publication Number
WO 02/23656 A1

(51) International Patent Classification⁷: H01M 8/02, 8/24

(21) International Application Number: PCT/US00/25391

(22) International Filing Date:
15 September 2000 (15.09.2000)

(25) Filing Language: English

(26) Publication Language: English

(71) Applicant: LYNNTECH, INC. [—/US]; 7610 Eastmark Drive, College Station, TX 77840 (US).

(72) Inventors: CISAR, Alan, J.; 15603 Juniper Hollow Way, Cypress, TX 77429 (US). MURPHY, Oliver, J.; 4213 Meadowbrook, Bryan, TX 77802 (US). CLARKE, Eric, T.; 401 Oak Street, Bryan, TX 77801 (US).

(74) Agent: STREETS, Jeffrey, L.; Streets & Steele, P.O. Box 1612, Cypress, TX 77410-1612 (US).

(81) Designated States (*national*): AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW.

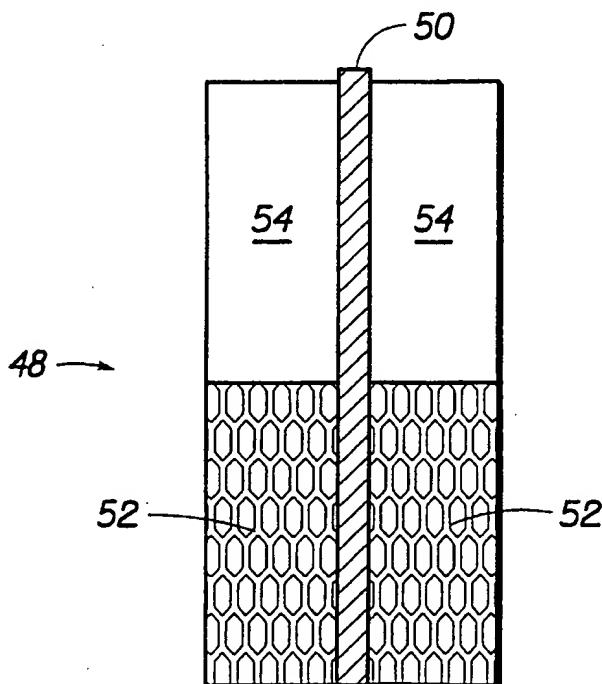
(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

Published:

— with international search report

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: BONDING ELECTROCHEMICAL CELL COMPONENTS



(57) Abstract: The invention provides a method for preparing subassemblies for an electrochemical cell or a stack of electrochemical cells, particularly a stack of fuel cells for the direct generation of electricity. The method includes bonding together two or more electrochemical cell components, such as plates, frames, flow fields, shims, gaskets, membranes and the like, to form subassemblies used to make an electrochemical cell stack. The bonding can be accomplished using either polymeric bonds (i.e., adhesives) where polymer and/or metal components are involved or metallurgical bonds (i.e., solder) where metal components are involved. The bonding provides tightly sealed cells and lower electronic contact resistances between components.

WO 02/23656 A1

BONDING ELECTROCHEMICAL CELL COMPONENTS

Field of the Invention

The present invention relates to electrochemical cells and methods for
5 assembling electrochemical cells.

Background of the Invention

Electrochemical cells utilizing a proton exchange membrane (PEM) can be
configured in cells stacks having bipolar separator plates between adjacent cells. These
bipolar separator plates are typically made from a variety of metals, such as titanium
10 and stainless steel, and non-metallic conductors, such as graphitic carbon. Bipolar
separator plates are typically fabricated by machining flow fields into a solid sheet of
the material. Alternatively, when carbonaceous conductive materials are used, the
precursor material is formed by injection molding and converted to the conductive
carbon form by high temperature firing under carefully controlled conditions. The flow
15 fields are made up of a series of channels or grooves that allow passage of gases and
liquids.

Figure 1 is a face view of a prior art bipolar separator plate 10 made from a
solid sheet of a conducting material. The central portion of the plate has a flow field 12
machined into its surface. The flow field may direct fluid flow across the surface of an
20 electrode in many patterns, but is illustrated here as parallel serpentine channels.
Around the perimeter of the flow field 12, the plate provides a plurality of bolt holes 14
for assembling and securing a cell stack, various manifolds 16 for communicating
fluids through the stack, and a flat surface 18 that allows the plate to be sealed against
adjacent components of the cell stack.

25 In addition to providing a fluid flowfield, a bipolar separator plate for use in
electrochemical cells must collect electrons liberated at one electrode (i.e., an anode),
conduct the electrons through the plate, and deliver electrons to the face of another
electrode (i.e., a cathode) on the opposing side of the plate. The bipolar plate shown in
Figure 1, collects and delivers electrons from electrodes of opposing cells through
30 contact between the electrodes and the ridges 20 remaining between the channels 22 in
the flowfield 12.

Figure 2 is a schematic view of a proton exchange membrane (PEM) electrochemical cell configured as a hydrogen-air fuel cell stack 30. This stack 30 comprises two identical fuel cells 32 each having a cathode 34, a PEM 36 and an anode 38. Flow fields 40 (shown schematically for clarity) are provided on either side of the bipolar separator plate 42, as well as on the internal faces of the endplates 44. Electrons liberated at the anodes 38 induce electronic current flow to the cathode 34 of an adjacent cell on the other side of the plate 42 and, in the case of the last anode of the stack (here the anode on the right of the page), through an external circuit 46. Electrons are then combined with protons and oxygen at the cathodes 34 to form water. The electrical potential of the fuel cell 30 is increased by adding more cells 32 to the stack.

While the foregoing methods are relatively straight forward, they have several disadvantages. First, the solid piece of graphite or metal used to fabricate the bipolar separator plate constrains the density of the final product to a density approximately the same as that of the original stock, thereby producing a very dense and heavy bipolar separator plate. Second, machining each piece from a solid starting blank requires relatively expensive machining processes, as opposed to less expensive molding, casting or stamping processes. When carbon components are used the molding step is inexpensive, however, the controlled sintering required to convert the precursor to the final product is slow and requires precise atmosphere and temperature control throughout the process.

Another important aspect in fabricating an electrochemical fuel cell is the number of joints and junctions created in the cell. Reduction of the number of joints and junctions can greatly improve the performance of a electrochemical cell stack, for example if fabricated from a stack of flat components, because there are fewer potential leak points and fewer electronic contact resistances. A fabrication process that provides an electrochemical cell with a minimum of joints and junctions would be highly desirable.

Assembling a PEM fuel cell stack using relatively flat components requires gas tight seals at each interface. Gaskets are typically used to create gas tight seals, however gaskets increase the number of parts that must be fabricated and aligned when the stack is assembled. A method and apparatus for forming gas tight bonds or seals at

the interfaces between the components of an electrochemical cell would obviate the need for several gaskets and produce a more efficient cell.

Therefore, there remains a need for an improved bipolar separator plate. It would be desirable if the bipolar separator plate were thin, light weight, and could support high current densities. It would be further desirable if the bipolar separator plate reduced the number of joints or junctions in the individual cells or a cell stack and reduced the need for gaskets. Furthermore, it would be useful if the structure of the bipolar separator plate allowed the introduction of other specific properties, such as water permeability and reactant gas impermeability.

10

Summary of the Invention

The present invention provides a method for preparing a subassembly for an electrochemical cell. The method includes aligning a subassembly having two or more electrochemical cell components with one or more bonding elements disposed between the two or more electrochemical cell components. The bonding elements have a melting point temperature that is lower than the melting point temperature of any one of the two or more electrochemical cell components. The subassembly is compressed and heated to a temperature that is between about the melting point temperature of the bonding element and about the lowest melting point temperature of the any one of the two or more electrochemical cell components. Preferably the temperature is less than 800°C, more preferably below 250°C. The subassembly is then allowed to cool.

The subassembly is preferably positioned into an electrochemical cell or an electrochemical cell stack. The two or more electrochemical cell components are preferably metal components selected from plates, shims, frames, flow fields or combinations thereof, such as stainless steel, titanium, nickel, nickel plated aluminum, nickel plated magnesium, or combinations thereof. The bonding element is preferably solder. The metal component is preferably dipped in a flux; then dipped in a bonding metal or solder, such as tin or a silver-tin alloy. The bonding metal, or solder can also be applied to the metal surface by electrodeposition or by various vacuum deposition techniques.

Light or easily oxidized metal components, such as those made from aluminum, magnesium, or alloys containing aluminum or magnesium are preferably coated with a

layer of a corrosion resistant transition metal prior to the dipping the metal component in the flux. Suitable corrosion resistant transition metals include but are not limited to cobalt, copper, silver, nickel, gold or combinations thereof. Nickel is the most commonly used metal for the corrosion resistant layer.

5 Alternatively, the two or more electrochemical cell components can be polymer components selected from frames, gaskets, membranes, shims, or combinations thereof where the bonding element is preferably an adhesive. The two or more electrochemical cell components may also comprise one or more metal components and one or more polymer components, where the bonding element is an adhesive.

10 The two or more electrochemical cell components can include a plate and a flow field. The subassembly preferably includes a bipolar plate and a frame. The bipolar plate preferably has two plates, a flow field and a frame. The frame and flow field are disposed between the two plates with the frame disposed around the flow field. The frame has channels in fluid communication with the flow field.

15 In another embodiment of the invention, there is provided a fluid cooled bipolar plate assembly having two electronically conducting plates having opposing faces, an electronically conducting flow field bonded in electronic communication with a substantial portion of the opposing faces of the plates, between the two electronically conducting plates, and a frame disposed around a perimeter of the electronically
20 conducting flow field and bonded between the two electronically conducting plates. The frame has channels for providing fluid communication between the flow field and a fluid source.

 Preferably, an electronically conducting cathode flow field and an electronically conducting anode flow field are bonded to opposing sides of the assembly. The
25 assembly is preferably bonded with an adhesive and/or solder.

 In yet another embodiment there is provided a bipolar plate for electrochemical cells having two or more porous, electrically conducting sheets selected from expanded metal mesh, woven metal mesh, metal foam, conducting polymer foam, porous conductive carbon material or combinations thereof. An electrically conducting gas
30 barrier is disposed in electrical contact between the sheets. A cell frame is disposed around a periphery of any one of the two or more porous electrically conducting sheets. The cell frame has at least one surface that is bonded to the gas barrier.

Preferably, the cell frame includes channels in fluid communication with the porous electrically conducting sheet. The cell frame can be metallic, where it is bonded to the gas barrier with a metallic bond. The metallic bond is preferably formed by soldering the cell frame to the gas barrier. Alternatively, the cell frame can be polymeric, where it is bonded to the gas barrier with a polymeric bond. Preferably the polymeric bond is produced by an adhesive.

Brief Description of the Drawings

So that the above recited features and advantages the present invention can be understood in detail, a more particular description of the invention, briefly summarized above, may be had by reference to the embodiments thereof which are illustrated in the appended drawings. It is to be noted, however, that the appended drawings illustrate only typical embodiments of this invention and are therefore not to be considered limiting of its scope, for the invention may admit to other equally effective embodiments.

Figure 1 is a face view of a typical metal separator plate with a serpentine flow field design.

Figure 2 is a schematic cross-section of a PEM fuel cell. The relative thickness of the components has been greatly exaggerated for clarity.

Figure 3 is a partial cross-section of a bipolar plate having a metal gas barrier with metal flow fields and polymer cell frames.

Figure 4 is a schematic drawing of the flat components used in the three dimensional structure of a fluid cooled bipolar plate.

Figure 5 is a graph showing the performance of a fuel cell stack of the present invention.

Detailed Description of the Preferred Embodiment

The present invention relates to components for use in electrochemical cells and methods for fabricating those components, including bipolar separator plates. More particularly, the present invention relates to a method for bonding adjacent components of an individual cell and/or adjacent cells of a stack. The bonding provides gas tight

seals which reduces or eliminates the need for certain gasket components and reduces or eliminates certain electronic contact resistances.

One aspect of the invention provides a bipolar plate for an electrochemical cell having an electrically conducting flow field, at least one gas barrier and a cell frame.

5 The components of the bipolar plate are bonded to each other to form gas tight bonds with either polymeric type bonds or metallic type bonds.

Another aspect of the present invention provides a method for bonding conductive portions of a bipolar plate. The conductive pieces are tinned or plated to coat them with a conductive metal and subsequently bonded together under pressure
10 and heat to form bonds between the parts.

The component parts of a bipolar plate or other member of an electrochemical cell may be categorized as being either those that must be conductive for the stack to function or those that are not required to be conductive. The flow fields and the gas barriers of the bipolar plate must be fabricated from a conductive material, such as a
15 metal or a conductive form of carbon, such as graphite. The cell frames and the sealing surfaces, also known as shims may be electronically insulating or conductive and can optionally be fabricated from various polymers, thereby resulting in a lower density component and significant overall weight savings for the cell or stack.

When individual components of a stack are fabricated from polymers and other
20 components are fabricated from metals, there are three possible types of interfaces present in a fuel cell stack, namely polymer-to-metal interfaces, polymer-to-polymer interfaces (such as if the membrane and electrode assemblies (M&E's) are bonded in place) and metal-to-metal interfaces. Each of these types of interfaces are bonded differently to form an electrochemical cell stack.

25 Figure 3 is a partial cross-section of a bipolar plate 48 having a metal gas barrier 50 with metal flow fields 52 and polymer cell frames 54, which essentially take the place of gaskets. The flow fields 52 are shown here as foamed metal, but may be fabricated from expanded metal, perforated metal, metal mesh screens or other porous conductive materials, as described in U.S. patent application serial number 08/787,271,
30 which is incorporated by reference in its entirety herein. The metal gas barrier 50 extends beyond the flow fields 52, preferably to the edge of the stack, with separate cell frames 54 on both the anode and cathode sides of the gas barrier 50. Flow fields 52 are

placed against the barrier 50, and inside the frames 54 as shown. Additional gasketing may be included between the barrier and the frame if desired.

Figure 4 is a schematic drawing of the components used in the three dimensional structure of a fluid cooled bipolar plate. A fluid cooled bipolar plate 55 is shown assembled from a series of substantially planar components including two cooling fluid barriers 60 and a cooling fluid frame 62, with an electronically conducting flow field 63 therein, similar to the flow field 52 of Figure 3. Optionally, the fluid cooled bipolar plate 55 may further include an anode cell frame 58, cathode cell frame 64 and sealing plates 56 (for contacting and securing a PEM or membrane and electrode (M&E) assembly).

This bipolar plate includes an internal cooling flow field 63 that allows passage of a cooling fluid for cooling the stack. The cooling fluid flows from a cooling fluid inlet manifold 65, through the cooling flow field 63 within the cooling fluid frame 62, and into a cooling fluid outlet manifold (not shown) generally similar to, but opposed from, the inlet manifold 65. The flow field 67 in the anode cell frame 58, the flow field 63 in the cooling fluid frame 62 and the flow field 69 in the cathode cell frame 64 may be made from any porous conductive material as described above.

Bonding Polymers-to-Metals

Bonding the polymer elements of a lightweight PEM fuel cell stack, such as the anode and cathode cell frames, to metallic elements, such as the gas barriers may be done with an adhesive that bonds to both surfaces to form a gas-tight seal that remains stable under typical cell operating conditions.

Four types of polymers were tested for fabricating components for PEM fuel cells (polycarbonate, polyethersulfone, polyetherimide, and polyimide). These polymers are representative of thermoplastics considered to be useful for this application. The polymer sheets were bonded to gold-plated titanium sheets using different adhesives to determine the combinations that produced adequate bonding for use in electrochemical cells. Gold plated titanium was chosen for these experiments because it is considered to be one of the most challenging materials, useful in conductive electrochemical cells components, to bond with a polymer.

A variety of adhesives were used to bond 0.032" (0.813 mm) polymer sheets to gold plated 0.007" (0.178 mm) titanium sheets. The test specimens were

approximately 2 centimeters wide and 2 centimeters long. The procedure involved preparing the surfaces, applying the adhesive, bonding, and curing the adhesive according to the individual manufacturers instructions. The bonded specimens were then exposed to water at 60 °C for 24 hours, after which they were examined to determine the quality of the bond. A representative group of adhesives showing reasonable bonding characteristics are included in Table I.

Table I. Adhesion Testing of Adhesively Bonded Polymer/ Metal Couples.

Polymer	Adhesive	Results
Polycarbonate	Acrylic-epoxy hybrid	Fair adhesion to both surfaces and fair cohesion
Polycarbonate	Butadiene	Good adhesion to both surfaces; fair cohesion
Polycarbonate	Acrylic	Fair adhesion to both surfaces; poor cohesion
Polycarbonate	Polyurethane	Bond fails at Ti surface when wet
Polycarbonate	Neoprene	Good adhesion and cohesion properties but pigment leaches into water
Polycarbonate	Acrylic w/rubber	Bond fails at Ti surface when wet
Polycarbonate	Heat Seal	Excellent adhesion and cohesion properties when wet
Polyethersulfone	Butadiene	Good adhesion to both surfaces; fair cohesion
Polyethersulfone	Polyurethane	Bond fails at Ti surface when wet
Polyethersulfone	Neoprene	Excellent adhesion and cohesion properties but pigment leaches into water
Polyetherimide	Butadiene	Good adhesion to both surfaces but fair cohesion
Polyetherimide	Polyurethane	Bond fails at Ti surface when wet
Polyetherimide	Neoprene	Excellent adhesion and cohesion properties but pigment leaches into water
Polyetherimide	Acrylic w/rubber	Bond fails at gold plated Ti surface when wet
Polyetherimide	Heat Seal	Bond fails at gold plated Ti surface when wet
Polyimide	Silicone	Bond fails at plastic surface
Polyimide	Pd/Ag Epoxy	Good adhesion and cohesion properties
Polyimide	Acrylic-epoxy hybrid	Bond fails at plastic surface

**Table II. Product and Manufacturer Identification for the Adhesives
Appearing in Table I**

Adhesive	Product Name	Manufacturer
Acrylic-epoxy hybrid	PLASTIC WELDER II	Devcon Inc., Danvers, MA
Butadiene	GOOP and E6000 STIK 'N SEAL	Eclectic Products, Carson, CA Loctite, Inc., Newington, CN
Acrylic	Acrylic 204 and Acrylic 406	Lord Adhesives, Erie, PA
Polyurethane	PLIOGRIP 7775L	Ashland Chemical, Columbus, OH
Neoprene	SCOTCH GRIP 1357	3M, St Paul, MN
Acrylic w/rubber	Acrylic 330	Loctite, Inc., Newington, CN
Heat Seal	Polyurethane 3218	Bemis Assoc. Inc., Shirley, MA
Silicone	748 Silicone Rubber Cement	Dow-Corning, Midland, MI
Pd/Ag Epoxy	Pd/Ag filled Epoxy	Epotek, Inc., Billerica, MA

This list is considered to be representative and in no way exhaustive of the types
5 of materials useful in carrying out the present invention.

Bonding Metals-to-Metals

Two types of metal-to-metal bonds are present in an electrochemical cell. There
are bonds that are critical to the conductive path of electrons, such as the bonding of
10 flow fields to gas barriers, and there are bonds that are critical to seal against fluid
leakage, such as the bonding of gas barriers to metal frames. With regard to the gas
tight bonds, it is beneficial, but typically not necessary, if the bond is also conductive.

Conductive bonds can be created between a variety of metals by soldering the
parts together. Soldering is a well known technique where a relatively low melting
15 metal is used to bond two components together that are fabricated from metals having
higher melting points than the low melting metal or solder. It is imperative that the low
melting metal wets the higher melting metals to achieve a good bond.

Conventional soldering for wire attachment for an electrical assembly involves
heating the parts to above the melting point of the solder, then applying a flux to

remove the oxide film on the metal and finally applying the solder, with the flux and the solder sometimes applied together. Soldering large areas requires a different process. In order to solder large flat areas, the part is first coated with the bond-forming metal or alloy by one of several methods. It can first be coated with flux and
5 then coated with solder in a process known as tinning. The flux can be applied in a variety of ways, however brushing and dipping are the most commonly used methods. Application of solder is typically accomplished by dipping the part in a container of molten solder. Specific approaches to tinning parts fabricated from stainless steel, nickel, titanium, magnesium, aluminum, and alloys containing aluminum and
10 magnesium are described below. The bond forming metal or alloy can also be plated onto the metal surface by either electrodeposition or electroless deposition, both of which are well known processes. Also, the bond forming metal or alloy can be deposited onto the metal surface using vacuum deposition techniques, such as evaporation, chemical vapor desposition and sputtering.

15 After depositing the bond forming metal by tinning or plating, the parts are bonded by clamping the flat surfaces to be bonded together and reheating the assembly to above the softening point of the solder. Both the clamping and the heating can be accomplished simultaneously through the use of a hot press.

The procedure for tinning stainless steel parts includes, dipping the part in an
20 acid flux solution; slowly dipping the part in molten solder; dipping the part in the acid flux a second time; and dipping the part in molten solder a second time. Preferably, the tinning procedure is carried out in an inert atmosphere. The clean stainless steel part is immersed in a water soluble flux, preferably an acid flux, and the excess solution is allowed to drain off. The part is then slowly lowered into the molten solder. It is
25 important that the part be immersed slowly, because the flux is only active for removing the oxide film while the flux is hot, and a slow immersion process leads to a longer exposure time and better oxide removal. A silver solder, such as eutectic tin-silver (96.5 Sn : 3.5 Ag) is preferred. In some cases, a single immersion in the molten solder is sufficient, but in other cases a second dipping is preferred. In the latter case,
30 after the part is removed from the solder, it is allowed to cool slightly to insure that the solder has solidified, and immersed in the flux a second time. The part is then removed from the flux and the excess solution allowed to drain off. Ideally, the part should be

sufficiently hot to boil the flux as it makes contact. In the last step, the part is slowly lowered into the molten solder and removed.

The first immersion in the solder covers most of the surface of the stainless steel part with a thin layer of solder, however occasionally there are areas where the flux did not remove all of the oxide film, that are poorly coated. The second dip in the flux with the part hot removes any remaining oxide film from the part and produces a shiny, mirror-like finish on the metal. It is important that all grease and superficial dirt be removed from the surface of the metal before soldering. Failure to clean and degrease the surface may leave a protective film on the oxide layer that prevents the flux from cleaning the surface, and prevents the solder from sticking. After tinning, the parts are preferably thoroughly washed in deionized water to remove all remaining traces of the flux. Because water soluble flux residues float on the surface of the molten solder, most parts will have traces of flux present on the tinned surface. Other types of flux may be used to carry out the tinning process.

The same tinning procedure works for titanium, aluminum, magnesium, and alloys containing these elements as well, if they are first plated with nickel, copper, or another metal that more readily accepts tinning. Precious or base metals may be used to plate the titanium.

The procedure for tinning titanium, aluminum, and magnesium containing parts includes plating the part with nickel, dipping the part in an acid flux solution, and dipping the part in molten solder. In order to nickel plate the part, the oxide film must be removed.

After the part has been nickel plated, it is coated with flux and dipped in molten solder to produce a shiny tinned finish using the same cycle and silver solder as is used with stainless steel. Generally, only a single immersion is required for each the flux and the solder with nickel-plated aluminum.

Both aluminum and titanium can be tinned directly if the proper combination of flux and solder are employed. With the use of a high viscosity liquid flux, aluminum and titanium can be directly tinned. In this process the part is first immersed in the flux, and then slowly immersed in molten solder, with a single cycle being sufficient. For these metals, a solder with a more active component is preferred, such as eutectic tin-zinc (91% Sn, 9% Zn, melting point 199°C).

It is important to note that for best results, all of the tinning operations described herein are preferably carried out in an oxygen free atmosphere, or an atmosphere with a substantially reduced oxygen content. It is not necessary, and indeed in most cases it is not possible, to exclude water, since water is a major component of many fluxes.

5 As long as a tin based solder is being used, it doesn't matter what alloying elements are present in the solder when forming the final bonds, the tinned parts can readily be bonded together. When solders with dissimilar compositions are used to tin metals to be bonded it is important to heat the clamped components to at least the melting point of the higher melting solder.

10 While the metallurgical bond produced by soldering metal components together is preferred, it is not the only approach to joining metal components. Adhesives, like those listed in Table II, can be used for some metal-to-metal bonds as well. In general, these adhesives are suitable for bonds where through-bond electrical conductivity is not required. Except for the Pd-Ag filled epoxy, none of the adhesives listed in Table II are
15 conducting. The conductive adhesive bond may be substituted for a soldered joint in producing an electrochemical cell or stack in accordance with the present invention.

 Still other metal joining processes applicable to this type of stack fabrication will be apparent to those skilled in the art of metal joining. These are also considered to be part of this invention when they are employed for the assembly of an
20 electrochemical cell stack in a manner essentially similar to that described here.

Bonding Polymers to Polymers

 There are many adhesives available which are suitable for bonding engineering thermoplastics to each other. However, bonding PEM membranes to other materials is
25 more difficult because perfluorinated membranes are resistant to adhesion by most adhesives. Dow-Corning 748™ adhesive (see line 8 in Table II) adheres well to the membrane and to many metal surfaces. While the peel strength, at about 1.3 kg per centimeter of bond, is inadequate for most structural purposes, it is more than adequate to produce a thin, fully adherent seal for use in an electrochemical cell.

30

Assembly Procedures

Assembling a fully bonded electrochemical cell stack includes several steps. First, all of the metal parts are coated with the bonding metal, including the anode flow field, cathode flow field, and cooling flow field; the gas barriers; and the frame around
5 the cooling flow field. These parts are then stacked into position, carefully positioned, and held in place using an alignment device or jig. The parts and the jig are then placed in a hot press to clamp the parts together, pressing the gas barriers firmly against the cooling frame and the flow fields firmly against the gas barriers. The press is heated to slightly above the melting point of the highest melting bonding metal present in order
10 to remelt the bonding metal. The bonded assembly is then allowed to cool in the press.

Sheets of hot melt adhesive are cut to match the size and shape of the anode and cathode cell frames. These are used to bond the cell frames to the cell shims through the use of the hot press. Those two piece subassemblies are bonded to the assembled bipolar plate by the same technique. All of the hot melt bonds can be made in a single
15 step, if desired.

An uncooled bipolar plate is somewhat easier to fabricate because it contains few parts. The flow fields are bonded to opposite sides of a gas barrier by soldering. The anode and cathode cell frames are attached to the gas barriers and the shims with hot melt adhesives in one or two pressing operations.

20 The following examples show some of the preferred embodiments of the present invention, however they are not to be considered limiting in any sense.

Example 1

This example shows the fabrication of the core of a liquid cooled bipolar plate.

25 Two titanium gas barriers were fabricated from 0.0045" (0.114 mm) metal sheet and plated with gold. A frame was fabricated from 0.032" (0.813 mm) polyether-sulfone sheet. A flow field was fabricated from three layers of expanded titanium, one with a thickness of about 0.030" (0.762 mm) at full expansion and two which were expanded and subsequently flattened back to their original 0.003" (0.076 mm)
30 thickness. All three sheets were spot welded together, with the 0.003" sheets welded to opposing faces of the 0.030" sheet, and the welded assembly was gold plated. This flow field was deliberately produced thicker than the frame to insure that the flow field,

which compresses like a spring, would put pressure on the gas barriers to ensure electrical contact with the barriers.

The faces of the polyethersulfone frame were coated with ECLECTIC E6000 adhesive. The frame was then pressed, by hand, against one gas barrier, with both parts
5 kept in alignment by the use of a positioning jig fabricated for this purpose and equipped with alignment pins to insure that all of the parts stay precisely in alignment. The flow field was placed in the cavity produced by the frame on the barrier, and the second gas barrier was pressed on top of the other parts. The assembly, still in the assembly jig, was placed in a hot press. The components were then pressed together
10 and cured under sufficient compression to keep the parts from moving at 60 °C for 24 hours. The result was a bipolar plate having a gas barrier with fluid access to the internal flow field suitable for passage of a cooling fluid for the purpose of removing waste heat from the fuel cell stack. In subsequent assembly steps, this subassembly was treated in the same manner as a simple gas barrier sheet.

15

Example 2

This example shows the bonding of two components with hot melt adhesive.

A face shim or sealing plate 56 was fabricated from 0.0045" (0.114 mm) titanium sheet, as illustrated in Figure 4. An anode cell frame was fabricated from
20 0.032" (0.813 mm) polyethersulfone sheet. The anode cell frame was used as a template to cut a sheet of hot melt adhesive (BEMIS number 3218). All three pieces were stacked in a positioning jig fabricated for this purpose and equipped with alignment pins to insure that all of the parts stay precisely in alignment with the adhesive between the metal shim and the polymer frame. The jig, with the parts inside
25 of it, was placed in a hot press, compressed to insure and maintain intimate contact between the sheet components, and heated to 150°C. (The temperature used for this step is a function of the adhesive used.) After several minutes, the press was cooled, and the subassembly removed.

This subassembly is suitable for use as the frame around the anode flow field
30 and the contact surface against the membrane portion of a membrane and electrode assembly (M&E) and used in assembling a fuel stack.

Example 3

This example shows the tinning of stainless steel. A gas barrier was fabricated from 0.010" (0.254 mm) stainless steel sheet. The gas barrier and the other items required to carry out the tinning operation (flux, solder, heater, tongs, temperature indicator, etc.) are placed in a glove box and the atmosphere of the box thoroughly purged with argon to prevent oxidation of freshly cleaned surfaces. The barrier is immersed in an acid type liquid flux such as LA-CO N-3 (LA-CO Industries, Chicago, IL). Excess liquid is allowed to drain-off stainless steel sheet, and the piece is slowly immersed in molten silver-tin eutectic solder (3.5 % Ag, 96.5 % Sn, melting point 221 °C, (Kester Solder, Des Plaines, IL). It is important to immerse the piece slowly, since the flux is only active and capable of dissolving the surface oxide layer on the metal over a limited temperature range, and a slow immersion insures an adequate exposure of the entire surface of the part to the flux at its active temperature, but before all of the salts active for dissolving the oxide film vaporize.

After immersion, the part is removed from the molten solder, with all parts of the surface which were stripped of oxide by the action of the flux being coated with solder. The part is allowed to cool until the solder solidifies before being re-immersed in the flux. It is preferable to have the part remain sufficiently hot to boil the liquid slightly as it contacts the surface of the metal. The part is removed from the flux and slowly re-immersed in the solder, with the second cycle serving to remove the oxide film from the parts of the surface which were not stripped the first time and coat those areas with solder. After the part is removed from the molten solder it is allowed to cool in the air until the solder solidifies. The part may then be laid down on a heat resistant surface or other support to finish cooling. The result is a mirror-like finish on the tinned part.

Example 4

This example shows the tinning of aluminum. A cooling cell frame for a fluid cooled bipolar separator plate was fabricated from 0.020" (0.508 mm) aluminum sheet and plated with nickel. The gas barrier and the other items required to carry out the tinning operation (flux, solder, heater, tongs, temperature indicator, etc.) were placed in a glove box and the atmosphere of the box was thoroughly purged with argon to

prevent oxidation of freshly cleaned surfaces. The barrier was immersed in an acid type liquid flux such as LA-CO N-3 (LA-CO Industries, Chicago, IL). Excess liquid was allowed to drain-off, and the piece was slowly immersed in molten silver-tin eutectic solder (3.5 % Ag, 96.5 % Sn) having a melting point of 221 °C (Kester Solder, Des Plaines, IL). It is important to immerse the piece slowly, since the flux is only active and capable of dissolving the surface oxide layer on the metal over a limited temperature range, and immersing the part slowly ensures that the entire surface of the part is adequately exposed to the flux at its active temperature, but before the volatile components are vaporized. After immersion, the part is removed from the molten solder with the surface completely coated with solder.

Example 5

This example shows the bonding of aluminum and stainless steel components to form the core of a liquid cooled bipolar plate using solder.

Two gas barriers and a flow field are tinned as described in Example 3. An aluminum cooling frame is tinned as described in Example 4. The components are stacked as shown in Figure 4 and placed in a hot press with a thermocouple in contact with the side of the frame and clamped firmly together. The press is heated until the thermocouple indicates that the load has reached and maintained a temperature of about 230 °C for about five minutes, at which time the power to the heaters is turned off and the entire assembly is allowed to cool while maintaining the assembly in a compressed state. The result is a water cooled gas barrier with fluid access to the internal flow field suitable for use as a component in a fuel cell stack with the specific purpose of removing waste heat from the stack. This subassembly was treated in the same manner as one of the simple gas barrier sheets in subsequent assembly steps.

Example 6

This example illustrates the performance of a fuel cell stack produced in accordance with the present invention.

The method of Example 1 was used to prepare a set of four water cooled bipolar plate cores sized for use in a fuel cell stack with an active area for 125 cm². Three uncooled bipolar plates were also used in the stack. The method of Example 2 was

used to prepare eight anode cell frames with shims and eight cathode cell frames with shims for a stack of that size. Additional components (M&E's, flow fields, gaskets, etc.,) were also prepared by conventional means. These were used to fabricate an eight cell PEM fuel cell stack. The stack was operated using hydrogen fuel with air as the oxidant. Figure 5 illustrates the performance of that eight-cell stack, 125 cm² per cell active area while operating at an M&E temperature of 62 - 64°C, a fuel gas pressure of 15 psig, both fuel and oxidant gases humidified to a dew point of 27°C, and air supplied at four-fold stoichiometry. The stack provided an output of 967 W/kg and 846 W/L for the repeating units of this stack which was determined to be an efficiency of 53.4% (at a potential of 0.651 V/cell).

While the foregoing is directed to preferred embodiments of the present invention, other and further embodiments of the invention may be devised without departing from the basic scope thereof, and the scope thereof is determined by the claims which follow.

What is claimed is:

- 1 1. A method for preparing a subassembly for an electrochemical cell, comprising:
2 aligning a subassembly having two or more electrochemical cell components
3 with one or more bonding elements disposed between the two or more electrochemical
4 cell components, wherein the bonding elements have a melting point temperature that is
5 lower than the melting point temperature of any one of the two or more electrochemical
6 cell components;
7 compressing the subassembly;
8 heating the subassembly to a temperature less than 800°C; and
9 allowing the subassembly to cool.
1
- 1 2. The method of claim 1, further comprising:
2 positioning the subassembly into an electrochemical cell.
1
- 1 3. The method of claim 1, further comprising:
2 positioning the subassembly into an electrochemical cell stack.
1
- 1 4. The method of claim 1, wherein the two or more electrochemical cell
2 components are metal components selected from plates, shims, frames, flow fields or
3 combinations thereof and the bonding element is a metal or metal alloy that melts at a
4 temperature below 500 °C.
1
- 1 5. The method of claim 4, wherein the metal components are selected from
2 stainless steel, nickel, aluminum, titanium, magnesium or combinations thereof.
1
- 1 6. The method of claim 4, further comprising:
2 dipping the metal component in a flux; then
3 dipping the metal component in a solder.
1
- 1 7. The method of claim 6, wherein the solder comprises a metal selected from tin
2 bismuth, lead, indium, and alloys thereof.
1

- 1 8. The method of claim 6, wherein the solder comprises silver and tin.
1
- 1 9. The method of claim 6, further comprising:
2 coating the metal component with a layer of a corrosion resistant transition
3 metal prior to the dipping the metal component in the flux.
1
- 1 10. The method of claim 4, wherein the bonding metal is applied to one of the metal
2 component surfaces by reductive deposition from a solvent.
1
- 1 11. The method of claim 4, wherein the bonding metal is applied to one of the metal
2 component surfaces by vacuum evaporation or sputtering.
1
- 1 12. The method of claim 10, wherein the solvent is selected from water or a water
2 based fluid.
1
- 1 13. The method of claim 10, wherein the deposited metal comprises a component
2 selected from tin, bismuth, lead, indium or an alloy thereof.
1
- 1 14. The method of claim 1, wherein the two or more electrochemical cell
2 components are polymer components selected from frames, gaskets, membranes,
3 shims, or combinations thereof and the bonding element is an adhesive.
1
- 1 15. The method of claim 1, wherein the two or more electrochemical cell
2 components comprise one or more metal components and one or more polymer
3 components, and wherein the bonding element is an adhesive.
1
- 1 16. The method of claim 1, wherein the two or more electrochemical cell
2 components include a plate and a flow field.
1
- 1 17. The method of claim 1, wherein the subassembly includes a bipolar plate.
1
- 1 18. The method of claim 17, wherein the subassembly further includes a frame.
1

1 19. The method of claim 17, wherein the bipolar plate comprises two plates, a flow
2 field and a frame, wherein the frame and flow field are disposed between the two plates
3 with the frame disposed around the flow field, and wherein the frame has channels in
4 fluid communication with the flow field.

5

1 20. A fluid cooled bipolar plate assembly comprising:
2 two electronically conducting plates having opposing faces;
3 an electronically conducting flow field bonded in electronic communication
4 with a substantial portion of the opposing faces of the plates, between the two
5 electronically conducting plates; and
6 a frame disposed around a perimeter of the electronically conducting flow field
7 and bonded between the two electronically conducting plates, wherein the frame has
8 channels for providing fluid communication between the flow field and a fluid source.

1

1 21. The assembly of claim 20, further comprising an electronically conducting
2 cathode flow field and an electronically conducting anode flow field bonded to
3 opposing sides of the assembly.

1

1 22. The assembly of claim 20, wherein the assembly is bonded with an adhesive.

1

1 23. The assembly of claim 20, wherein the assembly is bonded with solder.

1

1 24. A bipolar plate for electrochemical cells, comprising:
2 two or more porous, electrically conducting sheets selected from
3 expanded metal mesh, woven metal mesh, metal foam, conducting polymer foam,
4 porous conductive carbon material or combinations thereof;
5 an electrically conducting gas barrier disposed in electrical contact
6 between the sheets; and
7 a cell frame disposed around a periphery of any one of the two or more
8 porous electrically conducting sheets, wherein the cell frame has at least one
9 surface that is bonded to the gas barrier.

1

- 1 25. The bipolar plate of claim 24 wherein the cell frame includes channels in fluid
2 communication with the porous electrically conducting sheet,
1
- 1 26. The bipolar plate of claim 25 wherein the cell frame is metallic and is bonded to
2 the gas barrier with a metallic bond.
1
- 1 27. The bipolar plate of claim 25 wherein the cell frame is polymeric and is bonded
2 to the gas barrier with a polymeric bond.
1
- 1 28. The bipolar plate of claim 25 wherein the metallic bond is formed by soldering
2 the cell frame to the gas barrier.
1
- 1 29. The bipolar plate of claim 26 wherein the polymeric bond is produced by an
2 adhesive.

1/3

FIG. 1

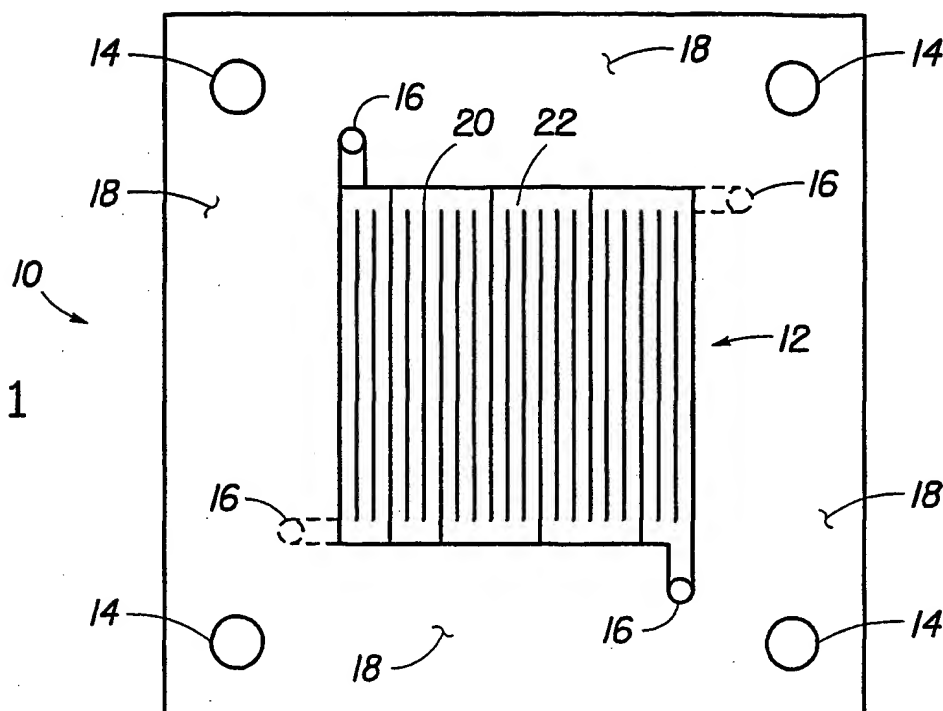
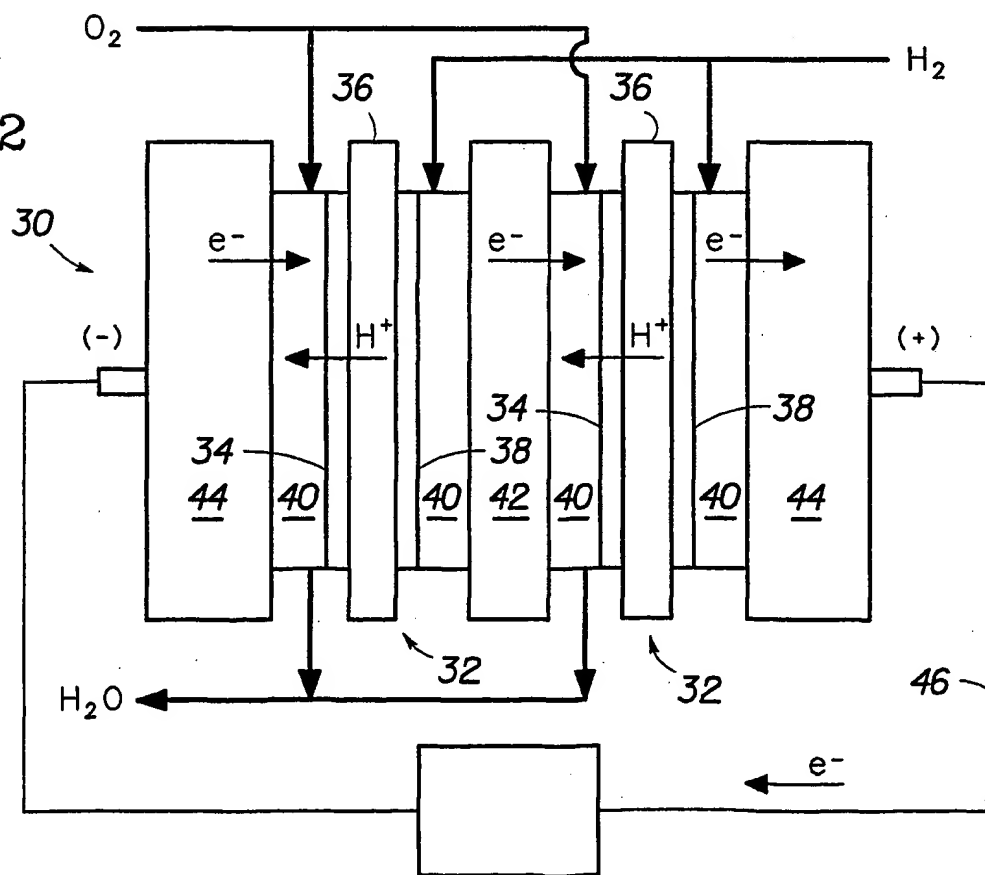


FIG. 2



2/3

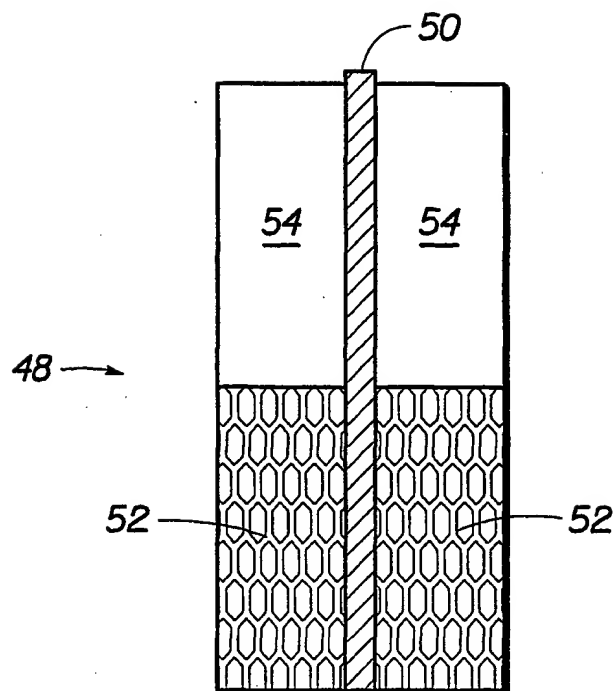


FIG. 3

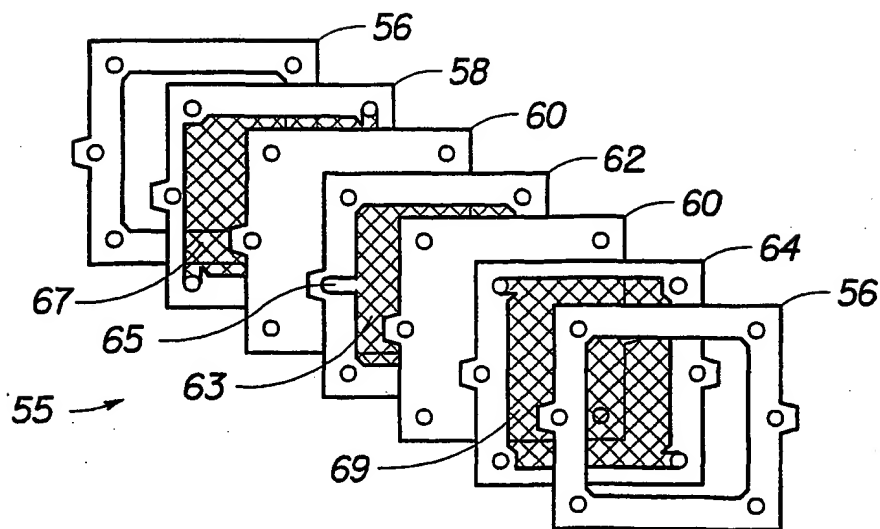


FIG. 4

3/3

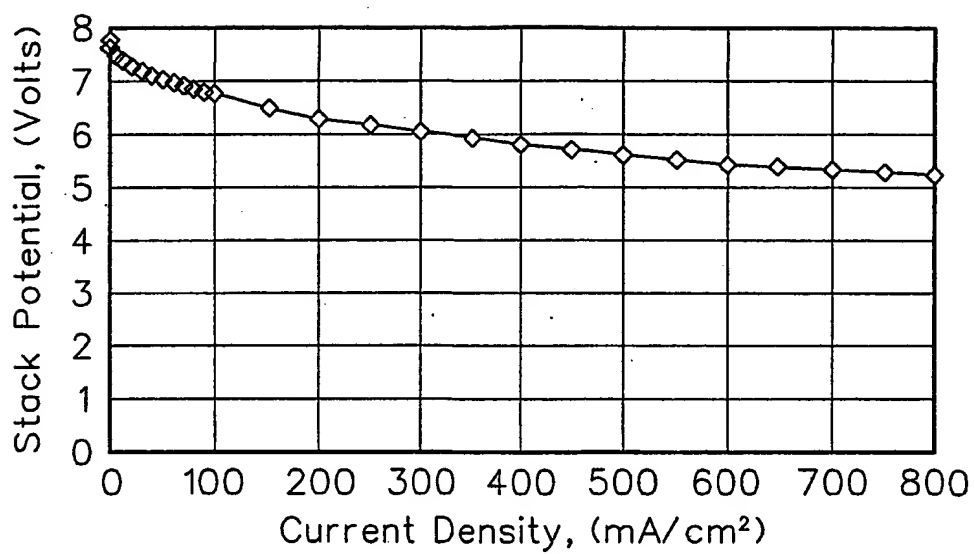


FIG. 5

Internal Application No
PC 17 00/25391

According to International Patent Classification (IPC) or to both national classification and IPC

Minimum documentation searched (classification system followed by classification symbols)

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
E	EP 1 089 369 A (CALSONIC KANSEI CORP) 4 April 2001 (2001-04-04) the whole document ----	1-23
X	US 3 717 505 A (UNKLE T ET AL) 20 February 1973 (1973-02-20) column 5, line 29 - line 54 ----	1-3, 14, 15
X	EP 1 009 051 A (GEN MOTORS CORP) 14 June 2000 (2000-06-14) page 4, paragraph 15 -page 5, paragraph 18 figures ----	1-3, 14, 15
X	US 3 126 302 A (DRUSHELLA R D) 24 March 1964 (1964-03-24) column 7, line 21 - line 45 ----	1-3, 14, 15

-/--

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- *A document member of the same patent family

Date of the actual completion of the international search

31 October 2001

Date of mailing of the international search report

07/11/2001

Name and mailing address of the ISA
European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Riba Vilanova, M

INTERNATIONAL SEARCH REPORT

 Int. Application No
 PCT/US 00/25391

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	FR 2 041 731 A (COMP GENERALE ELECTRICITE) 5 February 1971 (1971-02-05) page 2, line 27 -page 3, line 31 ---	1-3,14, 15
X	PATENT ABSTRACTS OF JAPAN vol. 009, no. 212 (E-339), 29 August 1985 (1985-08-29) & JP 60 074359 A (MITSUI ZOSEN KK), 26 April 1985 (1985-04-26) abstract ---	1-3,14, 15
X	DE 198 23 880 A (MOTOROLA INC) 10 December 1998 (1998-12-10) column 4, line 3 -column 5, line 24 ---	1-3,15, 17
X	DE 198 05 142 A (SIEMENS AG) 12 August 1999 (1999-08-12) column 2, line 61 -column 3, line 20 column 4, line 46 -column 5, line 21 figures ---	1-5
X	PATENT ABSTRACTS OF JAPAN vol. 017, no. 307 (E-1379), 11 June 1993 (1993-06-11) -& JP 05 029001 A (MITSUBISHI HEAVY IND LTD), 5 February 1993 (1993-02-05) abstract ---	1-5
X	PATENT ABSTRACTS OF JAPAN vol. 017, no. 460 (E-1419), 23 August 1993 (1993-08-23) -& JP 05 109415 A (MITSUBISHI HEAVY IND LTD), 30 April 1993 (1993-04-30) abstract ---	1-5
X	US 3 617 385 A (GRAY FOSTER L) 2 November 1971 (1971-11-02) column 5, line 13 - line 75 ---	1-3
X	WO 98 33221 A (LYNNTECH INC) 30 July 1998 (1998-07-30) claims figures ---	24,27,29
X	DE 40 16 157 A (ASEA BROWN BOVERI) 13 December 1990 (1990-12-13) the whole document ---	20,24
A	EP 0 884 794 A (PEUGEOT ;CITROEN SA (FR)) 16 December 1998 (1998-12-16) the whole document -----	24

INTERNATIONAL SEARCH REPORT

In al Application No
F ; 00/25391

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
EP 1089369	A	04-04-2001	JP 2001167782 A EP 1089369 A2	22-06-2001 04-04-2001
US 3717505	A	20-02-1973	NONE	
EP 1009051	A	14-06-2000	EP 1009051 A2 JP 2000173631 A	14-06-2000 23-06-2000
US 3126302	A	24-03-1964	NONE	
FR 2041731	A	05-02-1971	FR 2041731 A7	05-02-1971
JP 60074359	A	26-04-1985	NONE	
DE 19823880	A	10-12-1998	DE 19823880 A1 FR 2764122 A1 GB 2326017 A SE 9801942 A	10-12-1998 04-12-1998 09-12-1998 04-12-1998
DE 19805142	A	12-08-1999	DE 19805142 A1	12-08-1999
JP 05029001	A	05-02-1993	JP 2955069 B2	04-10-1999
JP 05109415	A	30-04-1993	NONE	
US 3617385	A	02-11-1971	NONE	
WO 9833221	A	30-07-1998	US 6146780 A AU 6648198 A EP 0963614 A2 WO 9833221 A2	14-11-2000 18-08-1998 15-12-1999 30-07-1998
DE 4016157	A	13-12-1990	DE 4016157 A1	13-12-1990
EP 0884794	A	16-12-1998	FR 2764443 A1 DE 69800462 D1 DE 69800462 T2 EP 0884794 A1 ES 2156022 T3 PT 884794 T US 6071636 A	11-12-1998 08-02-2001 28-06-2001 16-12-1998 01-06-2001 29-06-2001 06-06-2000